

IN THE UNITED STATES PATENT AND TRADE MARKS OFFICE

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Assignee: Her Majesty in right of Canada,  
as represented by the Minister of Natural  
Resources

ReIssue of  
U.S. Patent 6,073,478

Title: HYDROGEN SENSOR USING A SOLID HYDROGEN ION  
CONDUCTING ELECTROLYTE

Our File: 1004-75

November 27, 2001

The Commissioner of Patents  
and Trade Marks,  
Washington, D.C., 20231  
U.S.A.

Dear Sir:

This is an amendment to the application for  
reissue of U.S. Patent 6,073,478 issued June 13, 2000.

In the Abstract:

Please amend the abstract as follows:

A reliable gaseous hydrogen detection and  
measuring device which is simple, easy to use, does not  
require any reference gas supply, and which can be of  
reasonably rugged construction. The device utilizes a disc  
comprising a solid state ceramic hydronium conductor of the  
general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  together with a silver

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based electrode system on one side, and a catalytic noble metal electrode, such as platinum, on the other. By measurement of the output voltage across the electrodes, both the presence, and the amount, of hydrogen in a gaseous system can be determined.

In the Claims:

Please amend claim 1 as follows:

1. (Amended) A hydrogen detection device comprising in combination:

(a) a body of phosphate bonded ceramic electrolyte of the general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  having a first face spaced apart from a second face;

(b) a catalytic noble metal electrode layer on the first face of the body in electrical contact with the ceramic electrolyte;

(c) a silver ion modified layer on and in the second face of the body;

(d) a silver electrode in contact with the silver ion modified layer; and

(e) conductive leads electrically connected to each of the faces;

whereby the emf generated when the ceramic body is exposed to hydrogen gas is measured.

Please cancel claims 2 and 3.

Please amend claim 4 as follows:

4. (Amended) A hydrogen detection device according to Claim 1 wherein the first and second spaced apart faces on the ceramic body are substantially parallel to each other, and wherein the first and second faces are each substantially flat.

Please amend claim 7 as follows:

7. (Amended) A hydrogen detection device according to Claim 1 wherein the two conductive leads are attached to each of the noble metal layer and the silver electrode by means of a conductive cement.

Please amend claim 12 as follows:

12. (Amended) A method of detecting hydrogen in a gaseous system which comprises exposing a detection device comprising in combination:

(a) a body of phosphate bonded ceramic electrolyte of the general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  having a first face spaced apart from a second face;

(b) a catalytic noble metal layer on the first face of the body in electrical contact with the ceramic electrolyte;

(c) a silver ion modified layer on and in the second face of the body;

(d) a silver electrode in contact with the silver ion modified layer; and

(e) conductive leads electrically connected to each of the faces;

to the gaseous system, and measuring the emf generated across the two conductive leads.

Please amend claim 13 as follows:

13. (Amended) A method of measuring the concentration of hydrogen in a gaseous system which comprises:

(i) exposing a detection device comprising in combination:

(a) a body of ceramic electrolyte of the general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  having a first face spaced apart from a second face;

(b) a catalytic noble metal layer on the first face of the body in electrical contact with the ceramic electrolyte;

(c) a silver ion modified layer on and in the second face of the body;

(d) a silver electrode in contact with the silver ion modified layer; and

(e) conductive leads electrically connected to each of the faces;

to a plurality of gaseous systems each containing known amounts of hydrogen;

(ii) measuring the emf generated across the conductive leads by exposure to each gaseous system to provide a calibration curve for the device;

(iii) exposing the device to a gaseous system containing an unknown amount of hydrogen;

(iv) measuring the emf generated on exposure to gaseous system in step (iii); and

(v) comparing the emf measured in step (iv) with the calibration curve obtained in step (ii).

Please add claims 14-24 as follows:

14. A method according to Claim 12 wherein the noble metal is chosen from the group consisting of platinum and palladium.

15. A method according to Claim 14 wherein the noble metal is platinum.

16. A method according to Claim 14 wherein the noble metal is palladium.

17. A method according to Claim 13 wherein the noble metal is chosen from the group consisting of platinum and palladium.

18. A method according to Claim 17 wherein the noble metal is platinum.

19. A method according to Claim 17 wherein the noble metal is palladium.

20. A hydrogen detection device according to Claim 1 wherein the noble metal is chosen from the group consisting of platinum and palladium.

21. A hydrogen detection device according to Claim 20 wherein the noble metal is platinum.

22. A hydrogen detection device according to Claim 20 wherein the noble metal is palladium.

23. A hydrogen detection device according to Claim 1 wherein the first and second spaced apart faces on the ceramic body are substantially parallel to each other.

24. A hydrogen detection device according to Claim 1 wherein the first and second faces are each substantially flat.

Attached hereto is a marked up version of the changes to the claims in a paper entitled "Version with Markings to Show Changes Made".

#### R E M A R K S

The present amendment letter is being submitted within two years of the grant of original patent 6,073,478.

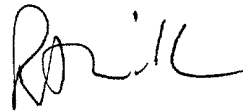
On receiving the issued patent, it was noted that the applicant disclosed palladium in the application and

failed to claim it. This error occurred through inadvertence without any fraudulent or deceptive intent.

Applicant has also taken this opportunity to correct typographical errors in the general formula in the abstract and in claim 13.

On reviewing the patent and discovering the error, the undersigned was requested to apply for reissue of the patent.

Respectfully submitted



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Version with Markings to Show Changes Made

The abstract has been amended as follows:

A reliable gaseous hydrogen detection and measuring device which is simple, easy to use, does not require any reference gas supply, and which can be of reasonably rugged construction. The device utilizes a disc comprising a solid state ceramic hydronium conductor of the general formula  ~~$\text{Na}(\text{H}_3\text{O})\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$~~   $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  together with a silver based electrode system on one side, and a catalytic noble metal electrode, such as platinum, on the other. By measurement of the output voltage across the electrodes, both the presence, and the amount, of hydrogen in a gaseous system can be determined.

Claim 1 has been amended as follows:

1. (Amended) A hydrogen detection device comprising in combination:

(a) a body of phosphate bonded ceramic electrolyte of the general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  having a first face spaced apart from a second face;

(b) ~~a layer of platinum on the first face of the body in electrical contact with the ceramic electrolyte a~~

catalytic noble metal electrode layer on the first face of the body in electrical contact with the ceramic electrolyte;

(c) a silver ion modified layer on and in the second face of the body;

(d) a silver electrode in contact with the silver ion modified layer; and

(e) conductive leads electrically connected to each of the faces;  
whereby the emf generated when the ceramic body is exposed to hydrogen gas is measured.

Claims 2 and 3 have been cancelled.

Claim 4 has been amended as follows:

4. (Amended) A hydrogen detection device according to Claim 1 wherein the first and second spaced apart faces on the ceramic body are substantially parallel to each other, and wherein the first and second faces are each substantially flat.

Claim 7 has been amended as follows:

7. (Amended) A hydrogen detection device according to Claim 1 wherein the two conductive leads are attached to each of the ~~platinum~~ noble metal layer and the silver electrode by means of a conductive cement.

Please amend claim 12 as follows:

12. (Amended) A method of detecting hydrogen in a gaseous system which comprises exposing a detection device comprising in combination:

(a) a body of phosphate bonded ceramic electrolyte of the general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  having a first face spaced apart from a second face;

(b) a ~~layer of platinum~~ catalytic noble metal layer on the first face of the body in electrical contact with the ceramic electrolyte;

(c) a silver ion modified layer on and in the second face of the body;

(d) a silver electrode in contact with the silver ion modified layer; and

(e) conductive leads electrically connected to each of the faces; to the gaseous system, and measuring the emf generated across the two conductive leads.

Claim 13 has been amended as follows:

13. (Amended) A method of measuring the concentration of hydrogen in a gaseous system which comprises:

(i) exposing a detection device comprising in combination:

(a) a body of ceramic electrolyte of the general formula  $\text{Na}(\text{H}_3\text{O})_x\text{Zr}_2\text{Si}_x\text{P}_{(3-x)}\text{O}_{12}$  having a first face spaced apart from a second face;

(b) a ~~layer of platinum~~ catalytic noble metal layer on the first face of the body in electrical contact with the ceramic electrolyte;

(c) a silver ion modified layer on and in the second face of the body;

(d) a silver electrode in contact with the silver ion modified layer; and

(e) conductive leads electrically connected to each of the faces; to a plurality of gaseous systems each containing known amounts of hydrogen;

(ii) measuring the emf generated across the conductive leads by exposure to each gaseous system to provide a calibration curve for the device;

(iii) exposing the device to a gaseous system containing an unknown amount of hydrogen;

(iv) measuring the emf generated on exposure to gaseous system in step (iii); and

(v) comparing the emf measured in step (iv) with the calibration curve obtained in step (ii).

Claims 14-24 have been added as follows:

14. A method according to Claim 12 wherein the noble metal is chosen from the group consisting of platinum and palladium.

15. A method according to Claim 14 wherein the noble metal is platinum.

16. A method according to Claim 14 wherein the noble metal is palladium.

17. A method according to Claim 13 wherein the noble metal is chosen from the group consisting of platinum and palladium.

18. A method according to Claim 17 wherein the noble metal is platinum.

19. A method according to Claim 17 wherein the noble metal is palladium.

20. A hydrogen detection device according to Claim 1 wherein the noble metal is chosen from the group consisting of platinum and palladium.

21. A hydrogen detection device according to Claim 20 wherein the noble metal is platinum.

22. A hydrogen detection device according to Claim 20 wherein the noble metal is palladium.

23. A hydrogen detection device according to Claim 1 wherein the first and second spaced apart faces on the ceramic body are substantially parallel to each other.

24. A hydrogen detection device according to Claim 1 wherein the first and second faces are each substantially flat.

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